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The purpose of the work described in this final report is to develop and exploit (exciplex-based) fluorescence diagnostics in order to investigate the heating and vaporization of hydrocarbon fuel droplets in both isolated and fuel spray environments. In the course of the investigations, experimental and computational techniques have been developed (a) for imaging (and correcting images) of an equatorial "slice" of sub-millimeter diameter droplets, (b) for exciplex fluorescence thermometry of optically thick droplets ("surface temperatures") and optically thin droplets ("volume averaged temperatures"), (c) for generation and/or study of droplets, particularly at pressures greater than 25 atmospheres, and (d) for two dimensional imaging of radiative lifetimes (equivalence ratio imaging).

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Applications of Laser-Induced Exciplex Fluorescence to Single Droplet and Fuel Spray Vaporization

Final Report

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July 12, 1991

U. S. Army Research Office

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I. STATEMENT OF THE PROBLEM STUDIED

The purpose of the work described in this final report is to develop and exploit (exciplex-based) fluorescence diagnostics in order to investigate the heating and vaporization of hydrocarbon fuel droplets in both isolated and fuel spray environments. In the course of the investigations, experimental and computational techniques have been developed (a) for imaging (and correcting images) of an equatorial "slice" of sub-millimeter diameter droplets, (b) for exciplex fluorescence thermometry of optically thick droplets ("surface temperatures") and optically thin droplets ("volume averaged temperatures"), (c) for generation and/or study of droplets, particularly at pressures up to 25 atmospheres, and (d) for two dimensional imaging of radiative lifetimes (equivalence ratio imaging).

It is hoped that the demonstration of the availability and utility of these new techniques will lead to will lead to their use in clarifying the basic physical processes involved in the heating and vaporization of sub-millimeter droplets.

L. A. Melton at the University of Texas at Dallas (UTD) has served as the principle investigator on this contract. Michael Winter at United Technologies Research Center (UTRC) is the principal scientist on the subcontract to UTRC. In so far as possible, individual results described in the text are labelled by the the source of the work.

II. SUMMARY OF MOST IMPORTANT RESULTS

A. Optical Properties of Droplets

1. Calculations of droplet absorption and emission

Melton at UTD developed three programs, based on ray tracing approximations: 1) TOTABS calculates the spatial distribution of excited states in a droplet which is irradiated by a plane wave, 2) FLUOR-EFF calculates the fraction of the light, which is emitted isotropically at a given point in a droplet, which enters the aperture of a collection lens, 3) COMBO calculates the convolution of the results of the previous two programs, i.e., the fraction of the incident light which is absorbed and then emitted into the lens aperture.

2. Calculation of undistorted images for "droplet slicing" experiments

The novel work carried out at UTRC on "droplet slicing" diagnostics, in which a thin laser sheet illuminates an equatorial plane of the droplet, has stimulated modeling

work at UTD to determine how to transform the "camera image" obtained in such an experiment into a "real image", appropriate for analysis by investigators interested in heat and mass transport within droplets, i.e., how to undo the distortions caused by refraction at the droplet/air interface.. Although this work is not yet complete, it appears that the real image can be accurately recovered out to fractional radii of 0.8-0.9.

B. "Droplet Slicing" Experiments

1. Internal circulation

At UTRC, Winter succeeded in "slicing" a 300 micron droplet with a 60 micron thick laser beam and thereby imaging a planar slice of the droplet (See section III(3)). In the first experiments, a decane droplet, doped with naphthalene and previously purged with nitrogen, was allowed to fall, at room temperature, into nitrogen or into air. The naphthalene fluorescence is strongly quenched by oxygen, and as oxygen from the air dissolves in the decane, the naphthalene fluorescence decreases. Internal circulation in the droplet carries liquid from the surface into the interior of the drop, and the oxygen rich regions appear as dark zones in the droplet fluorescence. The images are normalized against the pure nitrogen case in order to minimize the effects of refractive distortions. These experiments are the first time internal circulation has been observed in freely falling droplets this small.

The experiments described in the preceding paragraph established that internal circulation was present in the droplets; they did not, however establish whether the observed internal circulation was induced by the droplet formation process or by aerodynamic drag. Winter has pursued the design and testing levitation devices (allow formation process to decay before releasing the droplet to fall) and aerodynamic droplet generators (internal circulation induced by formation process opposite to internal circulation induced by aerodynamic forces during the fall) in order to carry out experiments in which will establish the role of aerodynamic forces on internal circulation.

At UTRC, Winter has routinely exploited his ability to produce droplets at high pressures (See section II.D.2) and has carried out "droplet slicing" imaging of O_2 diffusion/ convection into droplets at pressures from 1-25 atmospheres as a function of fall distance (i.e., time). At higher pressures, as expected, there is substantially more drag and substantially faster O_2 uptake.

By observing the motion of internal structures made visible by the addition of the oxygen-sensitive fluorescent dopants, Winter was able to demonstrate that the falling droplets also rotate.

2. "Droplet slicing" thermometry

At UTRC, Winter achieved temperature visualization in the equatorial plane of a droplet, under conditions almost equivalent to those used by Wells and Melton (See section II.C.1). A laser sheet approximately 15 microns thick sliced an 800 micron droplet, which contained a naphthalene/N,N,N'N'-tetramethyl-p-phenylene-diamine (TMPD) exciplex thermometry solution, and the resulting fluorescence was imaged at right angles onto a color CCD camera. For droplet temperature measurements, the thermometry is carried out by separating the RGB outputs of the CCD camera and dividing the blue intensity by the green intensity pixel by pixel, to obtain a ratio which can be calibrated against the liquid temperature. Thus, signal processing takes the place of complex optical systems with multiple mirrors and separate blue and green filters. As the ambient temperature increases, the intensity of the fluorescence in the blue increases, while that of the green decreases.

3. Proof of equatorial slicing

At UTRC, Winter carried out systematic imaging experiments in which the thin laser beam is displaced from the center of the droplet; the images vary dramatically, and thus it is possible for an experimentalist to determine from the image whether the droplet was sliced at its center. In particular, the full circumference of the droplet is illuminated only if the illumination is in an equatorial plane. These images were also compared with the results of a ray tracing simulation of the laser sheet intersecting droplets off center.

C. Temperature Measurements in Droplets

AT UTD, thermometry experiments were carried out with a laser beam which was wide relative to the droplet diameter.

1. "Near Surface" temperatures

At UTD, Wells and Melton measured the "near surface" temperatures of 225 micron decane droplets which had fallen 10 cm through nitrogen heated to 250 ^{o}C (See section III(5)). A very strongly absorbing solution of pyrene was used to limit excitation to approximately the outer 36 microns of the droplet, and analysis of the ratio of the monomer to excimer intensities allowed the determination of the droplet "surface temperature". The droplet diameter was

measured through photomicroscopy, and the droplet evaporation was determined to be less than 10% of the volume at an oven temperature of 250 ^{o}C. In the course of these measurements, the affects of optical density, laser power, oxygen quenching, thermal expansion, and evaporation on the accuracy of the measurements were assessed.

2. "Volume Averaged" Temperatures

AT UTD, Hanlon and Melton (See section III.(7)) extended thee work of Wells and Melton by (1) reconstructing the apparatus so that the temperature of the droplet could be measured at virtually any point in its 9.5 cm fall through heated nitrogen, (2) increasing the temperature of the ambient to 550 ^{o}C, and (3) using 1,3-di(1-pyrenyl)-propane (PYPYP) as the exciplex fluorescence thermometry dopant, which results in a concentration-independent thermometer useful up to 400 ^{o}C in the liquid. (At sufficiently low concentrations ($<10^{-4}$ M) the formation of intermolecular exciplexes is negligible; the intramolecular exciplexes remain.) However, in these measurements with 1×10^{-4} M PYPYP in hexadecane, the 280 micron diameter droplet was optically thin (absorbance along a diameter = 0.06, as opposed to Wells and Melton results where the absorbance along a diameter was 4). In each case studied (ambient temperatures from 250 ^{o}C to 550 ^{o}C), the droplet temperature appeared to rise slowly, jump sharply, and then continue a slow rise. The highest temperatures measured for the hexadecane droplets (normal boiling point = 287 ^{o}C) were approximately 200 ^{o}C. Video microscopy measurements showed no evaporation of the droplet.

It is highly unlikely that the temperature of the entire hexadecane droplet jumps 100-120 ^{o}C in 5 ms and then resumes a slow rise. It is much more likely that the excitation process and fluorescence collection process results in selective sampling of the temperature field within the droplet. This "partially selective optical sampling" (PSOS) process has been modeled using geometrical optics (See section II.A.1). The droplet acts as a lens, which focuses the incoming beam, and through convergence of rays reflected at the backside of the droplet, a "hot spot" is formed at a fractional radius of about 0.75. The programs developed in this work also calculate the fraction of the isotropically emitted fluorescence which is refracted into the detector aperture. Overall, 58% of the total fluorescence originates from the shell between fractional radii of 0.5-0.75, even though this shell accounts for only 30% of the droplet volume. Thus, it is probable that the "temperature jump" records the change of temperature of the selectively sampled portion of the droplet. If radial diffusion is the dominant mechanism for the heating of the interior of the droplets, then the jump records the passage of the thermal wave; if internal circulation is the dominant

mechanism, then the jump records the heating of the interior of an internal circulation vortex.

D. Novel Techniques for Monodisperse Droplets

1. Inexpensive, rugged droplet-on-demand generators

AT UTRC, droplet-on-demand generators, which produce droplets reproducibly, were produced without the use of high-voltage, expensive, tempermental piezoelectric crystals. The 25 mm diameter speaker from a set of personal stereo headphones, driven by a pulse of 2-20 volts, was used as the driver.

2. Droplets-on-demand at high pressure

AT UTRC, reliable production of uniformly-sized fuel droplets in the 300-1000 micron diameter range at pressures up to 400 psia (27 atm) was achieved. The droplet-on-demand generator operated in a 4 in. I.D x 8 in. high 60 atm pressure vessel which was fitted with quartz windows. Only modest adaptations of the droplet-on-demand generators which are currently used at one atmosphere were required; the most substantial modification thus far was the development of an annular constant head reservoir With the exception of experiments involving droplet imaging over fall distances of more than 2 cm, droplet experiments similar to those carried out at one atmosphere can be carried out at 20-30 atmospheres. In particular, Winter was able to perform "droplet slicing" experiments (illumination with a 20 micron thick laser sheet along an equatorial plane of the droplet) at elevated pressures.

Preliminary imaging experiments showed the dramatic effects of increased pressure on droplet drag coefficients and on the rotation of the droplets.

3. Generation of droplets for internal circulation experiments

At UTRC, Winter developed techniques for isolation of the effects of aerodynamic drag on the internal circulation patterns. The process of ejecting a droplet through the nozzle of standard droplet generators may induce internal circulation in the same direction as that caused by aerodynamic forces. However, when droplets are ejected into an electrostatic trap where they can be held long enough for any fluid motions due to the droplet formation process to damp out, the influence of aerodynamic forces on internal circulation can be unambiguously identified. Alternately, an aerodynamic droplet generator can be used to produce monodisperse droplets in the core of an axisymmetric jet. The gas flow around the end of a hypodermic needle strips

off droplets. Since the gas flow velocity is greater than the initial liquid velocity, any internal circulation due to the droplet formation process will be opposite to that produced by free fall. Both electrostatic levitators and aerodynamic droplet generators were obtained and tested successfully.

4. Avoidance of cavitation in droplet-on-demand generators

At UTRC, Winter developed techniques for improving the reproducibility of operation of droplet-on-demand generators. When a square wave is used to drive the droplet-on-demand the generator squeezes out a single droplet and then retracts quickly, sucking in air through the tip. To inhibit the inhalation of air, a reverse sawtooth wave was used, which provided fast contraction and slow relaxation. A circuit was designed to provide this type of electric pulse; stable and repeatable trajectories of mono-dispersed droplets were obtained.

E. Equivalence Ratio Imaging

At UTD, T-Q Ni and Melton carried out "proof-of-concept" experiments which indicate that two dimensional imaging of the fuel/oxygen equivalence ratio should be possible (See section III(6)). The technique is based on the capture of two two-dimensional images, the second delayed from the first by a few nanoseconds. With this information, it is possible to calculate both the fuel and oxygen concentrations, and hence, the equivalence ratio. The technique depends upon fast gating (a few nanoseconds) of the two intensifiers and upon having a fluorescent dopant which is very inefficiently quenched by oxygen. The U.S. Army Research Office has recently funded the purchase by UTD of the fast gated intensifiers. Fluoranthene was found to be a (unique?) dopant, whose probability of quenching by oxygen is approximately 0.01 per collision.

III. LIST OF PUBLICATIONS AND TECHNICAL REPORTS

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(2) S. J. Hale and L. A. Melton, "Absolute Quantum Yields for Exciplex Fluorescence", Applied Spectroscopy, **44**, 101 (1990).

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(7) T. R. Hanlon and L. A. Melton, "Exciplex Fluorescence Thermometry of Falling Hexadecane Droplets", Journal of Heat Transfer, submitted for publication.

IV. PARTICIPATING SCIENTIFIC PERSONNEL

1. Lynn A. Melton, principal investigator
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3. Stanley K. Nickle, graduate student, Doctor of Chemistry degree awarded May 1988

4. Steven D. Sample, technician

5. Mark R. Wells, graduate student, Doctor of Chemistry degree awarded August 1990

6. Tuqiang Ni, graduate student, Doctor of Chemistry degree awarded August 1990

7. Thomas R. Hanlon, graduate student, Doctor of Chemistry degree awarded May 1991

8. John Blair, technician

9. Susan M. Eshelman, graduate student, M. S. degree expected December 1991

10 Oscar Arce, graduate student, M.S. degree expected May 1992

V. REPORT OF INVENTIONS

None